

## Amendments to the Specification

Please amend the paragraph at page 2, lines 3-7, as follows:

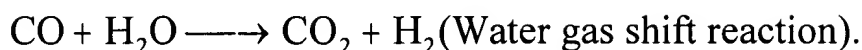
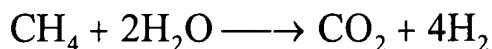
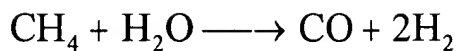
Molten chloride salt electrolytes (lithium chloride and potassium chloride) have been employed in electrochemical cells for the recovery of chlorine from hydrogen chloride gas. See, Yoshizawa, S. Et al.(1971) J. Appl. Electrochem. 245-251. U.S. patents 5,618,405 and 5,928,489 report the removal and ~~recovery~~ recovery of hydrogen halides from gas mixtures using molten halide salt electrolytes.

Please amend the paragraph at page 2, lines 8-17, as follows:

In electrochemical and fuel cells employing molten salt electrolytes, a porous electrolyte plate (or tile)[,] is made from a porous non-conducting matrix impregnated with the molten salt and positioned between an anode and a cathode. The porous matrix is typically made of a refractory, non-electron-conducting, inorganic material, such as lithium aluminate or lithium titanate. The electrolyte plate conducts or mediates ions between the anode and the cathode via the molten salt. The molten salt is selected for transport or mediation of a desired ion, e.g., a carbonate salt is used for mediation of a carbonate anion or a chloride salt is used for transport of chloride ion. The molten salt electrolyte plate does not conduct electrons. The anode and cathode of the electrochemical or fuel cells are electrically connected through an external circuit for electron transport.

Please amend the paragraph on page 3, lines 1-10, as follows:

Catalytic membrane reactors can also be employed for steam reforming of hydrocarbons. Steam reforming involves the following reactions ~~illustrate~~ illustrated with methane as the hydrocarbon:



U.S. patent 5,229,102 reports the production of CO<sub>2</sub> and H<sub>2</sub> by steam reforming of a hydrocarbon in a catalytic ceramic membrane reformer. In the membrane reactor, H<sub>2</sub> and CO<sub>2</sub> are generated by passing hydrocarbon and steam into the reactor zone in contact with a steam reforming catalyst, e.g., Ni metal promoted with alkali metal. Hydrogen is removed by permeation (or diffusion) through the membrane increasing the efficiency of the reaction.

Please amend the paragraph on page 3, lines 11-21 as follows:

Catalytic membrane reactors employing gas-impermeable, ion conducting membranes can, for example, be used for oxidation/reduction reactions. For example, oxygen or an oxygen-containing species (such as NO<sub>x</sub> or SO<sub>x</sub>) can be reduced at the reduction surface of a catalytic membrane to oxygen-containing anions which are transported across the membrane to an oxidation surface where they react to oxidize a selected reduced species. Materials used in the ~~mebranes~~ membranes in such a reactor conduct oxygen-containing anions. Provision must be made in such reactors for electron conduction to maintain charge neutrality permitting anion conduction through the membrane. Electron conduction has been achieved by the use of external circuits for current flow (U.S. patent 4,793,004). Electron conductivity has also been achieved by doping oxygen-anion conducting ceramic materials with metal ions to generate a material that conducts electrons and oxygen anions. See, U.S. patents 4,791,079 and 4,827,071.

Please amend the paragraph on page 4, lines 4-8, as follows:

The present invention relates to gas-impermeable mixed conducting membranes for use in a variety of catalytic membrane reactions and gas separation applications, which are formed by ~~impregnating~~ impregnating a porous electron-conducting matrix with a molten salt electrolyte. Ions mediated through these membranes facilitate gas separation and/or provide reactive species for the generation of desired value-added products in catalytic membrane reactors.

Please amend the paragraph on page 5, lines 2-13 as follows:

This invention relates to mixed-conducting membranes, i.e., membranes that conduct both ions and electrons, which behave as short-circuited electrochemical cells. The membrane comprises a porous electron-conducting matrix and a molten salt that conducts ions. The electron-conducting matrix is at least in part impregnated with molten salt to provide for ion transport through the membrane. The membrane comprises two external surfaces for contact, ~~respectivel~~ respectively, with reagent gas and reactant gas. Ions are transported from one external surface to the other external surface of the membrane. One or both of the external surfaces of the membrane can be catalytic. The external surfaces can be provided with adherent catalyst layers, or a three-dimensional catalyst can be provided in close proximity to one or both of the external membrane surfaces. Ions to be transported are formed at or near one external surface of the membrane in contact with reagent gas, transported through the membrane and released at the other external surface where they may react with reactant gas in contact with that surface.

Please amend the paragraph bridging pages 5 and 6 as follows:

More specifically, the membranes of this invention are useful for the generation of carbonate ion from carbon dioxide- and ~~oxygen-containing~~ oxygen-containing gas mixtures. The carbonate ion generated and mediated through the membrane is, in turn,

useful as a reagent ion for reaction with reactant gases to produce desired products and particularly for partial oxidation of reduced gases. Carbonate ion can be used for the partial oxidation of a variety of chemical species, including hydrocarbons. Using membranes of this invention, carbon dioxide can be removed from gas mixtures containing carbon dioxide and oxygen, particularly air, by reaction of carbon dioxide and oxygen to give carbonate ion at the membrane reducing surface. The carbonate ion produced is then transported through the membrane to the membrane oxidizing surface.

Please amend the paragraph on page 7, lines 6-22, as follows:

Pore size in the central layer of the electron-conducting matrix is typically on average less than about 1 micron and is preferably on average less than about 0.5 micron. Pore size in the external catalytic regions of the membrane is typically on average greater than about 1 micron and is preferably on average about 2-10 micron. The pores in the external regions of the membrane may have a bimodal distribution of sizes with the majority (greater than about 50%) of the pores sufficiently coarse that molten salt is not retained therein and with a minority (less than about 50%) of the pores sufficiently fine that molten salt is retained therein. In a preferred embodiment, up to about one-third of the pores in the external regions of the membrane are sufficiently fine in size to retain molten salt. The fine pores in the external regions of the membrane may be distributed non-uniformly with a higher amount of finer pores located in proximity to the central fine porosity region of the membrane. The membrane may, for example, be formed from a monolithic electron-conducting matrix having varying porosity with a central fine pore region, an intermediate region having a mixture of fine and coarse pores and an outer catalytic coarse pore region. Alternatively, the membrane may be formed ~~from multiple~~ from multiple layers of different porosity to achieve the desired pore size distribution and/or may be formed from layers of different electron-conducting materials of different porosity.

Please amend the paragraph bridging pages 7 and 8 as follows:

In a specific embodiment, the external regions of the porous membrane function to provide sites for catalytic oxidation and reduction reactions. One external catalytic region can provide a reducing surface when in contact with a gas containing carbon dioxide and oxygen (or other ion source gases) and the other (opposite) external catalytic region can ~~forms~~ form an oxidizing surface to which ions formed at the reducing surface are transported through the membrane. Reactions of anions, e.g., oxide anions, mediated through the membrane can occur at the oxidizing surface. For example, partial oxidation reactions effected by carbonate can occur at the oxidizing surface of the membrane when it is in contact with a gas containing a reactant gas or a reduced gas (e.g., methane or various hydrocarbons). Reactions of cations, e.g., ammonium ion, mediated through the membrane can occur at the reduction surface. For example, ammonium ion can be generated at the reduction surface of a membrane with concomitant reduction of a reactant gas, e.g. hydrogenation of a reactant gas.

Please amend the paragraph on page 8, lines 13-16, as follows:

The membranes of this invention conduct electrons and do not require external circuits to maintain charge neutrality when ions are mediated across the membrane. Thus, membranes of this invention do not require collector plates or similar electron-conducting ~~electron-conducting~~ elements that are employed in molten salt, electrochemical and fuel cells.

Please amend the paragraph on page 8, lines 17-24, as follows:

Metals useful for forming the electrolyte matrix are transition metals or mixtures thereof. Preferred metals for the porous matrix are nickel and mixtures of nickel with other transition metals. A preferred metal mixture is a mixture of nickel and chromium. Regions of the porous metal matrix of the membrane may react with components of the molten salt or gas components during operation (e.g., during exposure to reactant or

reagent gases). For example, regions of the metal matrix in contact with oxygen (or air) may be converted into metal oxide and portions of the metal matrix may react with the molten salt, e.g., the metal ~~matrix~~ matrix may become lithiated when Li molten salts are employed.

Please amend the paragraph on page 9, lines 3-14, as follows:

Molten salt electrolytes are selected for transport of a selected ion, e.g., molten carbonate salts are employed for carbonate ion transport. Membranes of this invention can be selected for transport of either anions, such as  $\text{CO}_3^{2-}$ , or cations, such as  $\text{NH}_4^+$ . The molten carbonate used in membranes of this invention is preferably a carbonate of an alkali metal or a mixture of alkali metal carbonates. More preferred molten carbonates are lithium carbonates, sodium carbonates, potassium carbonates or mixtures thereof. Molten halides can be employed for halide ion mediation, ~~mediation~~, e.g., alkaline halides, such as lithium halide or potassium halide or alkaline metal halides, such as lithium aluminum halides. Molten sulfate salts and nitrate salts can be used for sulfate and nitrate ion mediation, respectively. Molten phosphate salts can be used for phosphate ion mediation. Molten ammonium salts can be employed for ammonium cation mediation. Preferred molten salts for use in the membranes of this invention have melting points below about  $500^\circ\text{C}$ .

Please amend the paragraph on page 9, lines 15-28, as follows:

Gas-impermeable membranes of this invention can optionally be provided with adherent catalyst layers at either or both external surfaces to promote desired reactions. For example, adherent catalyst layers can be provided at the oxidation surface, the reduction surface or both to facilitate desired reactions. For applications to partial oxidation by oxide ions, an oxidation catalyst layer can be provided on the oxidation surface of the membrane. Particularly in applications to synthesis gas production at the oxidation surface of membranes of this invention, a reforming catalyst, particularly a steam reforming catalyst layer can be provided at the oxidation surface of the

membrane. A variety of reforming catalysts are known in the art including various metals (e.g., Ni, Ni-based alloys (e.g., Ni-Al or Ni-Cr), Co, Pt, Rh, Ru, Pd and mixtures thereof or noble metals catalysts) on various supports, including, e.g., alkaline metal oxides, alkaline earth metal oxides, silica, titania, zirconia, yttria, and mixtures thereof. Ni (about 5-about 10 weight%) on alumina is a ~~perferred~~ preferred catalyst. Catalysts can also be provided as a separate three-dimensional catalyst (e.g., particles or granules) in close proximity (including in contact with) one or both of the external surfaces of the membrane.

Please amend the paragraph on page 10, lines 1-22, as follows:

A catalytic membrane reactor of this invention comprises a mixed conducting membrane of this invention separating a reagent zone (where the ion to be transported is formed) and a reactant zone (where the ion transported is released for reaction). In a specific embodiment, the catalytic membrane reactor comprises an oxidation zone in contact with the oxidation surface of the membrane and a reduction zone in contact with the reducing surface of the membrane. In one embodiment where the reactive ions are anions, the reduction zone receives a gas mixture containing source gases for the reactive ion to be generated (i.e., a reagent gas) and the oxidation zone receives the reactant gas (e.g., a reduced gas). In another embodiment where the reactive ions are cations, the oxidation zone receives a gas mixture containing source gases for the reactive ion to be generated (i.e., a reagent gas) and the reduction zone receives the reactant gas (i.e., an oxidized gas). A reactor is also provided with appropriate gas inlets and gas outlets (e.g., a gas manifold) for gas handling, including source and reactant gas delivery and product gas collection. The reactor can also be provided with appropriate heating elements to initially heat the membrane to operating temperature in the range of about 500°C to about 800°C. The membrane is heated to a temperature that is at least sufficiently high to melt the electrolyte salt in the membrane. Operating temperature may be increased above the salt melting point to ensure efficient operation of the membrane and to optimize oxidation and/or reduction reactions. Operating temperature of the membrane may be, at least in part, maintained by heat released

from reactions occurring at the membrane surfaces. The reactor can be formed by positioning a membrane with appropriate seals between two chambers having appropriate gas inlets and outlets. A reactor contains one or more membranes of the invention and may be comprised of a stack of membranes separated by zones for introduction of gases ~~and or catalyst~~ and/or catalysts.

Please amend the paragraph bridging pages 10 and 11:

In a preferred embodiment, the catalytic membrane reactor of this invention is provided with a three-dimensional catalyst in the reagent zone, the reactant zone, or both of the reactor. In a specific embodiment, a three-dimensional catalyst can be provided in the oxidation zone or reduction zone of a reactor in close contact with the oxidation surface or the reduction surface, respectively, of the membrane. In a specific embodiment, for application to the generation of synthesis gas using carbonate mediating membranes, a three-dimensional reforming catalyst is provided in the oxidation zone of the reactor in contact with the oxidation surface of the membrane. For example, a steam reforming catalyst, as is known in the art, can be provided as a three-dimensional ~~catalys~~ catalyst, e.g., as particles or pellets, in the oxidation zone of the reactor in contact with the oxidation surface of the membrane to facilitate synthesis gas production in the catalytic membrane reactor. A reactor of this invention may incorporate both an adherent catalyst layer as well as a three-dimensional catalyst.

Please amend the paragraph bridging pages 13-14, as follows:

The membranes of this invention are mixed-conducting membranes which conduct ions, such as carbonate, as well as electrons. For example, carbonate ions formed at the reduction surface are transported through the membrane to the oxidation surface and electrons are concomitantly transported through the membrane to the reduction surface to maintain electronic neutrality in the membrane. Alternatively, cations formed at the oxidation surface are transported to the reduction surface and electrons are transported to maintain electronic neutrality. The membrane is formed of



a porous electron-conducting matrix at least partially impregnated with an ion-conducting molten salt. The amount of molten salt in the porous matrix is sufficient to provide for transport of a selected ion from one external membrane surface through the membrane to the other external surface. A molten carbonate salt is used, for example, to transport carbonate ions. The mixed-conducting membranes of this invention can be adapted for transport of various anionic species, such as halide ions, including  $F^-$ ,  $Cl^-$  and  $Br^-$ , and various oxide ions, including phosphate ion ( $PO_4^{3-}$ ), sulfate ion ( $SO_4^{2-}$ ) and nitrate ion [ $NO_3^-$ ]  $NO_3^-$  by appropriate choice of molten salt. i.e., anions of the molten salt comprise the anion to be transported. The mixed-conducting membranes of this invention can be adapted for transport of various cations, e.g., ammonium ion ( $NH_4^+$ ), again by appropriate choice of molten salt, i.e., cations of the molten salt comprise the cation to be transported.

Please amend the paragraph bridging pages 14-15 as follows:

In the membranes of this invention electron transport is provided by the electron-conducting matrix. The distribution of molten electrolyte within the electron-conducting matrix of the membrane is important to provide required ion paths through the membrane. Molten salt electrolyte should be available at the reaction sites, while not unnecessarily interfering with access of the gaseous reactants to and egress of gaseous products from the reaction sites. In preferred membrane structures, the resistance to ion flow between the catalytic surfaces (e.g., oxidation and reduction surfaces) of the membrane should be minimized. For example, the width of the central fine porosity region can be adjusted to increase ion flow, maintain gas-impermeability of the membrane and provide suitable mechanical strength for the membrane. The electrolyte distribution in the membranes of this invention is controlled by controlling pore size in the membrane matrix and by controlling the amount of molten salt added to the membrane matrix. Ideally the central region of the membrane, which separates the catalytic regions of the membrane, should contain as large a fraction of electrolyte as possible and the ionic path between the catalytic regions should be as short as is practical. The pores in the external regions of the membrane allow access of gases to

reaction sites and facilitate three phase interfaces (ions, electrons and reagents or reactants) to facilitate desired membrane reactions. In a preferred embodiment, at least about [one third] one-third of the pores in the external membrane regions contain some level of molten salt electrolyte.

Please amend the paragraph on page 16, lines 5-11, as follows:

Optimal pore size and pore size distribution in a given membrane can depend upon the type of electron-conducting matrix employed and the type of molten salt employed. Methods are known in the art ~~formaking~~ for making porous materials suitable for use as electron-conducting matrices in the membranes of this invention. Methods are also known in the art for controlling or adjusting average pore size in a variety of materials, including ceramics. For example, methods employed to prepare porous electrodes for ~~used~~ use in fuel cells can be employed or readily adapted in preparing the porous electron-conducting matrices of this invention.

Please amend the paragraph bridging pages 16-17 as follows:

Preferred molten salts for use in the membranes of this invention to transport anions are alkali metal salts, alkaline earth metal salts or mixtures thereof, including alkali metal carbonates, alkaline earth metal carbonates and mixtures thereof, alkali metal phosphate, alkaline earth metal phosphates and mixtures thereof and alkali metal nitrates, alkaline ~~earth~~ earth metal nitrates and mixtures thereof. For applications to carbonate ion generation and transport, alkali carbonates are employed. Molten carbonate electrolytes include, among others, mixtures of lithium and potassium carbonate, mixtures of lithium and sodium carbonate and mixtures of lithium, sodium and potassium carbonate. For example, mixtures of 65-40 mol%  $\text{Li}_2\text{CO}_3$  and 35 to 60 mol%  $\text{K}_2\text{CO}_3$  or  $\text{Na}_2\text{CO}_3$ , and preferably a 62/38 mol% mixture of  $\text{Li}_2\text{CO}_3$  and  $\text{K}_2\text{CO}_3$  or a 52/48 mol% mixture of  $\text{Li}_2\text{CO}_3$  and  $\text{Na}_2\text{CO}_3$  are useful in molten carbonate membranes of this invention. Ternary mixtures of  $\text{Li}_2\text{CO}_3$ ,  $\text{K}_2\text{CO}_3$ , and  $\text{Na}_2\text{CO}_3$ , particularly those in which  $\text{Li}_2\text{CO}_3$  is about 40 to about 65 mol% of the mixture, and

more particularly the mixture  $\text{Li}_2\text{CO}_3:\text{K}_2\text{CO}_3:\text{Na}_2\text{CO}_3$  (43.5:31.5:25 mol%) are useful in molten carbonate membranes of this invention. For applications to halide ion transport, lithium ~~halide~~ halide, potassium halide, lithium aluminum halide and mixtures thereof can be used, for example.

Please amend the paragraph bridging pages 19-20 as follows:

Figures 1A and B illustrate exemplary membranes of this invention. Fig. 1A illustrates a monolithic porous membrane **1** having a central fine pore region **3** in which substantially all of the fine pores **5** are substantially filled with molten salt, e.g., molten alkali or ~~alkaline~~ alkaline earth metal carbonate. Pores **6** generally increase in size in the external catalytic regions **7** and **8** of the membrane, which provide the reducing surface **9** and oxidizing surface **10**, respectively, of the membrane. The central region [3] **3** is illustrated in Fig. 1A as having an irregular shape. The central region may have a more regular or uniform shape. Alternatively, the membrane may be formed from an electron-conducting matrix having a high concentration of fine pores at the center with a uniformly decreasing concentration of fine pores and a uniformly increasing concentration of coarser pores as the distance from the center increases.

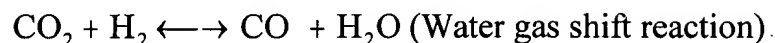
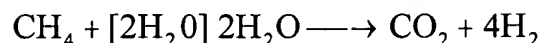
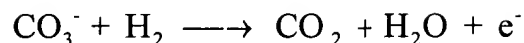
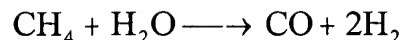
Please amend the paragraph bridging pages 20-21 as follows:

Fig. 2 illustrates an exemplary membrane reactor **30** of this invention. The reactor can be employed for carbon dioxide separation as carbonate ion and reaction of carbonate with a reactant gas in the oxidation zone of the reactor, e.g., partial oxidation reactions of the reactant gas. The reactor of Fig. 2 is illustrated for application of the production of synthesis gas by reaction of carbonate with a mixture of methane or natural gas in the presence of steam at least in part by steam reforming. The reactor has a reduction zone **31** and an oxidation zone **32** separated by the mixed conducting molten salt membrane **1**. Gas inlets **33** and **34** and gas outlets **35** and **36** are provided for the oxidation and reduction zones of the reactor. The oxidation surface **10** of the membrane is in contact with the oxidation zone and the reducing surface **9** of the

membrane is in contact with the reduction zone. The membrane is typically sealed in place in the reactor with a gas-impermeable sealing material **37**. The oxidation surface of the membrane is preferably provided with an adherent catalyst **38** that promotes ~~parital~~ partial oxidation. The reduction surface can optionally be provided with a reduction catalyst **39**. The reactor is illustrated with an optional three-dimensional catalyst in the oxidation zone **29**. In this ~~casse~~ case, the three-dimensional catalyst is illustrated as a reforming catalyst. The reactor is provided with a heater and temperature measurement and/or control for heating the membrane (not shown) to operating temperature and maintaining desired operating temperatures. A gas manifold (not shown) may be provided for control of the flow of gas or gases or for providing for mixing of gases in the oxidation and/or reduction zone. Reagent depleted gases, e.g., CO<sub>2</sub>-depleted gas, exiting the reduction zone may be recycled back into the reactor for further reaction or passed into another reactor. Product gases exiting the oxidation zone that contain unreacted reactant gases, such as methane, may be recycled back into the oxidation zone or passed on to another reactor for further reaction. Synthesis gas product exiting the reactor may be passed into another reactor for a variety of applications that are well known in the art.

Please amend the paragraph on page 21, lines 3-15, as follows:

Gas containing CO<sub>2</sub> and oxygen (reagent gas) is introduced into the reduction zone of the reactor. Carbonate ion is formed at the reducing surface of the membrane and transported to the oxidation surface of the membrane. Reactant gas containing a reduced gas, such as hydrogen, methane, ~~snatural~~ natural gas or other light hydrocarbons (or mixtures thereof) is introduced into the oxidation zone where it reacts with carbonate ion. Partially oxidized products are formed and exit the reactor. In particular, when methane and steam are introduced into the oxidation zone and react with carbonate, a mixture of CO and H<sub>2</sub>, synthesis gas, is produced. It is ~~beleived~~ believed that the following reactions occur:



In addition,  $\text{CO}_3^-$  can react directly to oxidize the hydrocarbon to CO and hydrogen.

Please amend the paragraph on page 21, lines 16-24, as follows:

Reagent gas (or ion-source gas) is a gas having one or more components at least one of which is used to generate selected ions at one external surface of the membrane. Ions may be generated by the reaction of one or more ~~reagent~~ reagent gas components at the membrane external surface (or in the external regions of the membrane). Reagent gases include sources of ammonium, carbonate, sulfate, halide, phosphate and nitrate ions among others. A source of carbonate can be  $\text{CO}_2$  and  $\text{O}_2$ . A source of sulfate can be  $\text{SO}_2$  and  $\text{O}_2$ . A source of nitrate can be  $\text{NO}_x$  and  $\text{O}_2$ . Various volatile halides can provide sources of halide ions. A source of ammonium can be ammonia and steam. A reagent gas can be an oxidized gas or an oxygen-containing gas.

Please amend the paragraph on page 22, lines 13-21, as follows:

Reactant gases can ~~include~~ include "reduced gases" which have at least one component that is capable of being oxidized at the oxidation surface of a reactor of this invention, for example by carbonate ion. Reactant gas components include, but are not limited to methane, natural gas (whose major component is methane), gaseous hydrocarbons including light hydrocarbons (as this term is defined in the chemical arts), alkanes, alkene, alkynes, and aromatic compounds, partially oxidized hydrocarbons such as methanol, higher alcohols (ethanol, etc.), aldehydes, ketones, epoxides and

may include organic environmental pollutants. Reactant gases include mixtures of reactant gas components, mixtures of such components with inert gases, or mixtures of such components with CO, CO<sub>2</sub> or H<sub>2</sub>O.

Please amend the paragraph on page 23, lines 9-25, as follows:

Fig. 3A illustrates an element of a stack assembly membrane reactor which can contain a plurality of membranes **1** of this invention. This reactor can be employed for gas separation and/or reaction of ions transported ~~through~~ through the membrane. The reactor **40** is shown with one membrane **1** in place in the stack assembly **41**. The reactor stack assembly typically contains a plurality of membranes **1** inserted into spaced slots **42**. Gas manifolds (as illustrated in exploded views in the figure) are provided for introduction and exit of gases from the reactor. The reactor is illustrated for production of synthesis gas and has a manifold **43** for introduction of gas containing CO<sub>2</sub> and O<sub>2</sub>, having a plurality of channels (not shown) and appropriately spaced inlets **44** to direct the gas mixture into reduction zones of the reactor. A gas outlet manifold **45** for depleted-CO<sub>2</sub> gas is also provided with a plurality of outlets **46** from each oxidation zone and appropriate channels **47** to direct the gas to exit the reactor **48**. A gas manifold **49** for introduction of methane (natural gas or other hydrocarbon) and steam, if desired, is also provided through appropriate channels (as illustrated at **47** for the outlet manifold ) with a plurality of appropriately spaced inlets **50** to direct the reactant gas into oxidation zones of the reactor. A plurality of product gas outlets **51** to conduct product gases (via channels that are not shown) to the exit **52** of the reactor are also provided. ~~Gas-impermeably~~ Gas-impermeable seals (not shown) are formed between the assembly frame **53** and each membrane.

Please amend the paragraph on page 24, lines 21-24, as follows:

Membranes of this invention are typically operated at temperatures between about 500°C[-] to about 800°C, preferably between about 550°C [-] to about 750°C and more preferably at about 650°C. Reactors employing the membranes of this invention

are preferably operated at 1 atm pressure, but may be operated at higher or lower pressures, if desirable.

Please amend the paragraph on page 25, lines 7-22, as follows:

Materials employed in the external or outer layers (8 and 7, in Fig. 1A and 22 and 23, in Fig. 1B) of the molten salt membranes (1) can function as catalysts for desired oxidation or reduction reactions. For example, coarse porous nickel or coarse porous nickel oxide in outer membrane layers can function for catalysis. The oxidation surface, the reduction surface or both of the molten salt membranes of this invention can be provided with adherent catalyst layers (e.g., a catalyst coating on the membrane surface or surfaces.) Specifically, the membranes of Figs[.], 1A or 1B can be provided with adherent catalysts. Catalysts that facilitate desired oxidation reactions can be provided at the oxidation surface, while catalysts that facilitate desired reduction reactions can be provided at the reduction surface. For applications to synthesis gas production, adherent reforming catalysts can be provided on the oxidation surface of the membrane. Reforming ~~catalyst~~ catalysts useful in this invention include, among others, Ni/Al<sub>2</sub>O<sub>3</sub>, doped LaMnO<sub>3</sub>, MnO<sub>2</sub> and doped LaNiO<sub>3</sub>, as are known in the art. Reforming catalysts useful in this invention also include, among others, various metals (e.g., Ni, Ni-based alloys (e.g., Ni-Al or Ni-Cr), Co, Pt, Rh, Ru, Pd and mixtures thereof or noble metals catalysts) on various supports, including e.g., alkaline metal oxides, alkaline earth metal oxides, silica, titania, zirconia, yttria, and mixtures thereof.

Please amend the paragraph on page 26, lines 17-19, as follows:

Adherent ~~catalyst~~ catalysts of this invention can be selected, for example, from mixed ionic and electronic conducting ceramics, cermet catalysts, platinum group of metals (Pt, Pd, Rh, or Ir) on oxide supports, or reforming catalysts as noted above.

Please amend the paragraph on page 27, lines 1-6, as follows:

Catalyst layers or coatings can be introduced [on to] onto membrane surfaces by a variety of methods. For example, a slurry of the catalyst powder in an organic solvent can be prepared and coated on to the membrane surface. The thickness of the layer or coating can be adjusted by varying the amount of slurry coated on the membrane or by adjusting the amount of catalyst in the slurry. The coated membrane is annealed at an appropriately high temperature to remove residual solvent.

Please amend the paragraph bridging pages 27-28 as follows:

The three-dimensional catalyst is selected, as is the adherent layer, to promote the desired oxidation or reduction reaction. An oxidation catalyst can be a metal on an inert oxide catalyst, such as Ni on  $\text{Al}_2\text{O}_3$  or other inert support. Alternatively, an oxidation catalyst can be a metal supported on a mixed ionic and electronic conducting material, such as Ni on  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ . The metal can be present in such catalysts from about 1wt% to about 50wt%. The catalyst in the packed-bed can comprise the same (or different) catalytic material as any adherent catalyst layer and can be chosen dependent upon application and reaction conditions from the catalysts listed in Tables 1 and 2 that are incorporated by reference from U.S. patent application 09/960,182, filed October 29, 1997 and published PCT application WO98/23051.

Please amend the paragraph on page 28, lines 13-18, as follows:

Figure 4 illustrates an exemplary membrane of this invention provided with an adherent catalyst layer (**64**). The membrane is illustrated as having a molten carbonate matrix for use in selective separation of carbon dioxide and oxygen as carbonate ion and subsequent reaction of the carbonate with methane for formation of synthesis gas. The membrane is illustrated as having external regions ~~22~~and 22 and 23 formed of coarse porous material. A steam reforming catalyst, such as Ni supported on alumina, can be provided as the adherent catalyst layer.



Please amend the paragraph on page 29, lines 14-15, as follows:

The following exemplary reactions can be carried out in reactors of this invention employing mixed conducting ~~membrane~~ membranes as described herein:

Please amend the paragraph bridging pages 29-30 as follows:

Alkenes, including those containing more than one double bond can be carboxylated with carbonate anion to yield mono or diacids. Alternatively, alkenes can be reacted with carbonate to yield alkyl carbonates. Dienes, such as butadiene, can be reacted with carbonate to yield cyclic lactones. Organic halides and ketones can be converted into acids by reaction with carbonate. Vinyl carbamates can be prepared by reaction of carbonate with a mixture of an alkyne and a secondary amine. Carbamate esters can be prepared by reaction of carbonate with primary [of] or secondary amines.